

Hysteresis and Anisotropic Magnetoresistance in Antiferromagnetic $Nd_{2-x}Ce_xCuO_4$

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The out-of-plane resistivity (ρ_c) and magnetoresistivity (MR) are studied in antiferromagnetic (AF) $Nd_{2-x}Ce_xCuO_4$ single crystals, which have three types of noncollinear antiferromagnetic spin structures. The apparent signatures are observed in $\rho_c(T)$ measured at the zero-field and 14 T at the spin structure transitions, giving a definite evidence for the itinerant electrons directly coupled to the localized spins. One of striking feature is an anisotropy of the MR with a fourfold symmetry upon rotating the external field (B) within ab plane in the different phases, while twofold symmetry at spin reorientation transition temperatures. The intriguing thermal hysteresis in $\rho_c(T, B)$ and magnetic hysteresis in MR are observed at spin reorientation transition temperatures.

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High- T_c superconductivity occurs in cuprates when doping introduces sufficient holes or electrons into the CuO_2 planes. It is generally believed that the pairing necessary for superconductivity involves the interplay between the doped charges and the AF spin correlation. In this sense, the study of lightly doped, insulating AF state is important because the density of the carriers can be sufficiently low that the interaction between them is small relative to their interaction with the Cu^{2+} spins. Many intriguing and anomalous phenomena show up in lightly doped AF cuprates due to the strong coupling between charges and magnetic order of the Cu^{2+} spins[1, 2, 3, 4].

In the hole doped cuprates, the Neel order is rapidly suppressed by doped hole, resulting in a "spin-glass" state [5] and a strong tendency to form spin-charge textures or "stripes" [6]. However, the long-range AF order in electron-doped $Nd_{2-x}Ce_xCuO_4$ persists to much larger x (≥ 0.12) [7], and coexists with superconductivity for even the optimal doping material ($x=0.15$) with $T_c=25$ K [8]. In addition, the Cu^{2+} spins order in an AF collinear structure for the parent compounds (such as: La_2CuO_4 and $YBa_2Cu_3O_6$) of hole-doped cuprates [9, 10], while in AF noncollinear structure for that of electron-doped cuprates: Pr_2CuO_4 and Nd_2CuO_4 [11, 12]. All spins point either parallel or antiparallel to a single direction in AF collinear structure, while the spins in adjacent layers are orthogonal in AF noncollinear structure. Magnetic-field induced a transition from noncollinear to collinear spin arrangement in adjacent CuO_2 planes for lightly electron-doped $Pr_{1.3-x}La_{0.7}Ce_xCuO_4$ with $x=0.01$ crystals affects significantly both the in-plane and out-of-plane resistivity [4]. In Nd_2CuO_4 , the Cu^{2+} spins order in three phases with two different AF noncollinear spin structures and experience two reorientation phase transitions [11, 13, 14, 15] as shown in Fig.1. Such reorientation phase transition is absent in Pr_2CuO_4 [16].

Magnetoresistance (MR) provides new insight into the coupling between the charges and the background mag-

netism. Previous experiments [1, 3, 4] have demonstrated that out-of-plane resistivity is sensitive to the interlayer magnetic order of the Cu^{2+} spins. This is particularly valuable because, as shown in this work, the spin-flop or flip transition occurs at fields in which magnetization measurements are difficult. In this letter, we systematically studied out-of-plane MR and angular dependence of out-of-plane MR for lightly doped $Nd_{2-x}Ce_xCuO_4$. It is found that $\rho_c(T)$ is surprisingly sensitive to the spin reorientation, giving a definite evidence for the itinerant electrons directly coupled to the localized spins. A thermal hysteresis in $\rho_c(T)$ at field and magnetic hysteresis in MR are observed. Another striking feature is the MR anisotropy with a fourfold symmetry in different AF spin structures, while with a twofold symmetry at the spin reorientation temperatures.

The NCCO single crystals were grown by flux method over a wide range of Ce concentration $0 \leq x \leq 0.13$. The actual Ce concentration was determined by inductively coupled plasma spectrometry (ICP) analysis experiments, and by the energy-dispersive x-ray analy-

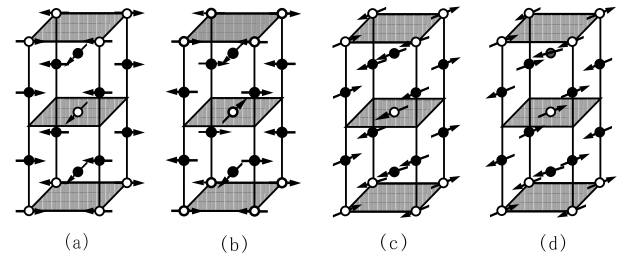


FIG. 1: Spin structure models for the AF noncollinear structure at zero field and the relative orientation of spins for the AF collinear structure at high field. (a) Noncollinear Phase I ($75 < T < 275$ K) and Phase III ($T < 30$ K); (b) Phase II ($30 < T < 75$ K); (c) Collinear Phase (type-I and III) induced by the field along the [110] from Phase I and III; (d) Collinear Phase (type-II) from Phase II. Here the open circles are Cu ions and the solid ones Nd ions.

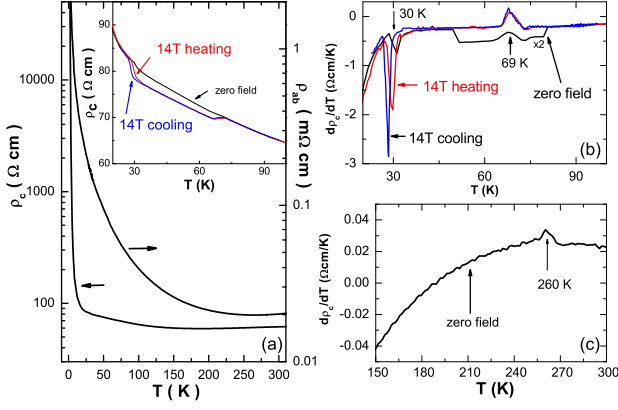


FIG. 2: (a) In-plane and out-of-plane resistivity as a function of temperature for $Nd_{1.975}Ce_{0.025}CuO_4$ crystal. (b)-(c) Temperature derivative of out-of-plane resistivity. In (b), the zero field data are multiplied by 2 between 50 to 80 K.

sis (EDX) using a scanning electron microscopy, respectively. All samples were annealed in flowing helium with purity of 99.999% for 10 hours at 900 °C to remove the interstitial oxygen. The resistivity and magnetoresistance were performed in Quantum Design PPMS systems.

The magnetic properties in R_2CuO_4 ($R=Nd, Pr$) are mainly dependent on the coupling between Cu and R magnetic subsystem which exhibits a large single-ion anisotropy [16]. Unlike Pr_2CuO_4 , in Nd_2CuO_4 the spin reorientation transition takes place due to a competition between various interplanar interactions which arises because of the rapid temperature dependence of the Nd moment below about 100 K [16]. The Cu spins first order in the noncollinear AF structure [phase I: Fig.1a] below $T_{N1}=275$ K. On further cooling, the Cu spins reorder into the noncollinear structure at $T_{N2}=75$ K [phase II: Fig.1b], and at $T_{N3}=30$ K the Cu spins experience another reorientation into phase III which has the same noncollinear order as phase I [11, 17]. As shown in Fig.1, the Cu and Nd moments along c-axis are parallel in phase I and III, while antiparallel in phase II [18]. A magnetic field applied within ab planes induces a transition from the noncollinear to collinear AF structure with a spin flop. Fig.1(c) and (d) show the collinear AF structures transformed from the noncollinear structures shown in Fig.1(a) and (b) at the B applied along [110] direction.

Figure 2a shows temperature dependence of in-plane (ρ_{ab}) and out-of-plane (ρ_c) resistivity for the crystal $Nd_{1.975}Ce_{0.025}CuO_4$. $\rho_{ab}(T)$ shows the insulating behavior in the whole temperature range, while $\rho_c(T)$ shows a weak metallic behavior above 130 K, and a weak insulating behavior with decreasing temperature, and a diverging below 30 K. At a first glance, no anomaly is detected at T_{N1} , T_{N2} and T_{N3} in the zero field $\rho_{ab}(T)$ and $\rho_c(T)$. However, a derivative plot for out-of-plane resistivity shown in Fig.1(b) and Fig(c) helps to observe the anomalies at the different T_N . As shown in Fig.1(b) and 1(c), the clear peaks are observed at $T_{N2}\sim 69$ K and

$T_{N3}\sim 30$ K for the spin reorientation transition, while a weak peak shows up at about 260 K for the AF order. Compared to Nd_2CuO_4 , the T_{N1} and T_{N2} slightly decreases. It suggests that doping of Ce suppresses the AF order and spin reorientation at T_{N2} , while does not affect the T_{N3} remarkably. At T_{N2} and T_{N3} , the width of the spin reorientation transition is very narrow (less than 10 K), indicating the high quality of our crystals. It should be pointed out that no anomaly is observed in ab-plane resistivity, even in its derivative. It suggests a strong coupling between the charge and spin degree of freedom, and that out-of-plane resistivity is more sensitive to the spin structure than in-plane resistivity. It should be pointed out that the anomalies shown in $d\rho_c/dT$ can be observed only for the $Nd_{2-x}Ce_xCuO_4$ crystals with $x<0.06$. In order to investigate the effect of collinear AF structure transition on $\rho_c(T)$, the $\rho_c(T)$ is measured at the B of 14 T along [110] direction in the heating and cooling process. As shown in the inset of Fig.2a, a remarkable feature is observed in $\rho_c(T)$ at T_{N2} and T_{N3} , and the transition from the type-I collinear structure to the type-II leads to a decrease in ρ_c . Very sharp peaks show up in a derivative plot ($\rho_c(T)/dT$) at T_{N2} and T_{N3} , respectively. The peak position at T_{N2} remains unchanged, while at T_{N3} obviously shifts to low temperature. A intriguing feature is that a hysteresis behavior at 14 T is clearly observed at T_{N3} and the peak temperature difference between heating and cooling process is about 1.5 K, while a hysteresis behavior can be ignored at T_{N2} . These results give the definite evidence for the itinerant electrons directly coupled to the localized spins. The similar hysteresis behavior cannot be observed at T_{N2} . Therefore, the hysteresis is induced by the external field.

Figure 3(a) shows the isothermal MR for $x=0.025$ with the B along Cu-O-Cu and Cu-Cu direction at 20 K and 40 K, respectively. The MR behavior shown in Fig.3(a) is similar to that observed in $Pr_{1.3-x}La_{0.7}Ce_xCuO_4$ with $x=0.01$ crystals by Lavrov et al[4]. As explained by Lavrov et al., the MR behavior arises from the spin origin and is closely related to the noncollinear-collinear transition induced by B. The steplike increase of resistivity corresponds to the noncollinear-collinear transition with increasing the field up to critical field B_c , above the B_c (in collinear structure) the MR shows different behavior. As shown in Fig.3(a), the Cu-Cu direction is easy axis in the collinear spin structure with relatively small B_c . Figure 3(b)-(e) shows the isothermal MR at 50 K with B applied within ab-plane Cu-Cu direction for the $Nd_{2-x}Ce_xCuO_4$ crystals with $x=0.025, 0.033, 0.06$ and 0.13 . An intriguing result is that magnetic hysteresis of MR is observed in the for $x=0.025, 0.033$ and 0.06 crystals. The magnetic field dependence of isothermal MR shows two branches. The branch of larger MR is obtained with field-cooled (FC) process, that is, the B of 14 T or -14 T is applied within ab-plane at 290 K, then the sample is cooled to 50 K with B, and the isothermal

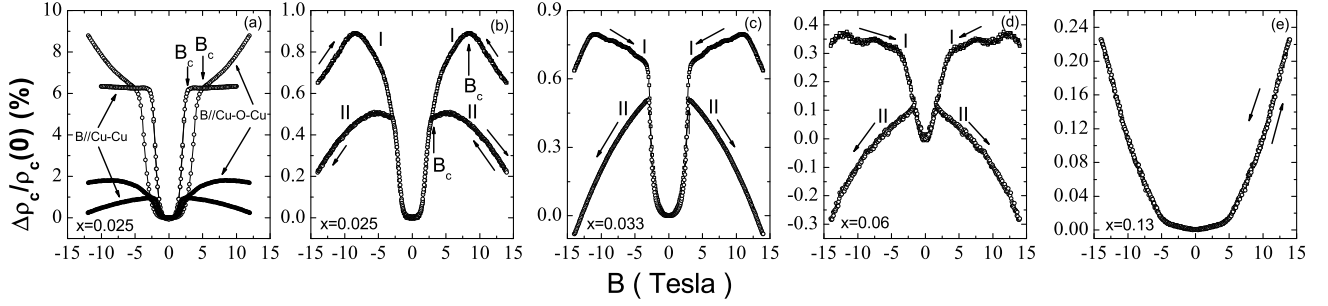


FIG. 3: (a) The isothermal MR for $x=0.025$ with the B along Cu-O-Cu and Cu-Cu direction at 20 K (open) and 40 K (solid), respectively; (b)-(e) The isothermal MR as a function of the B at 50 K in FC and ZFC process with B||Cu-Cu direction for the $Nd_{2-x}Ce_xCuO_4$ crystals with $x=0.025, 0.033, 0.06$ and 0.13 , respectively.

MR is measured with decreasing B. The branch of the smaller MR is got with increasing B from zero to 14 T, then with decreasing B the MR shows the same behavior as shown in Fig.3(b). The isothermal MR shows the same behavior as the smaller MR branch in zero-field cooled (ZFC) process. It suggests that the isothermal MR is strongly dependent on the B applied history. In Pr_2CuO_4 , no similar magnetic hysteresis of the MR is observed. While the difference of their magnetic structure is the absence of the spin reorientation in Pr_2CuO_4 . Therefore, the unique feature of the magnetic hysteresis in MR is closely related to the spin reorientation. As pointed out by Sachidanandam et al.[16] the spin reorientation transition originates from the competition between various interplanar interactions because of the rapid temperature dependence of the Nd moment below 100 K. It is possible that the magnetic field has an effect on various interplanar interactions. So the different effect of the field on the interplanar interactions exists in the different collinear spin structure, and leads to the different MR behavior between the ZFC and FC process. No magnetic hysteresis observed in ab-plane MR supports this explanation. No magnetic hysteresis is observed in Fig.3(e) for the $x=0.13$ crystal. This may be due to two possibilities: (1) antiferromagnetic order is completely suppressed by doping, or the T_{N1} is below 50 K; (2) the spin reorientation temperature T_{N2} is suppressed to be less than 50 K with doping, so that no spin orientation occurs above 50 K as the case of Pr_2CuO_4 . There exist two important differences between the MR branch I and II. First is the MR behavior above B_c and the sign of the anomalous MR, which is always positive for branch I, while negative at high fields for branch II. Second, the critical field B_c for the noncollinear to collinear spin structure transition is larger in branch I than that in branch II. Which could originate from the effect of the B on the interplanar interactions in the FC process enhances the critical field for the noncollinear-collinear spin transition. It should be pointed out that the hysteresis in MR is not observed below T_{N3} , and only can be observed at temperature between T_{N2} and T_{N3} .

In order to make out effect of spin reorientation transition on the MR, the MR as a function of angle at

different temperatures is studied. The angular dependence of the MR was determined by rotating the sample under a fixed field of 12 T within ab-plane. Figure 4 shows the evolution of the MR with angle between B and [100] (Cu-O-Cu) direction for the 0.025 crystal. The MR is always positive for all temperatures. A striking feature is that the MR shows a strong anisotropy with fourfold symmetry in different AF phases, while twofold symmetry around T_{N2} and T_{N3} . The similar anisotropy with *d-wave-like* symmetry has been observed in $Pr_{1.3-x}La_{0.7}Ce_xCuO_4$ with $x=0.01$ crystal [4]. The fascinating MR oscillations shown in Fig.4 has been explained to arise from the relative orientation of spins with respect to the crystal axes because the spin structure always stays collinear at high fields. The total energy does not change due to the interplane pseudo-dipolar interactions when the spin sublattices of the adjacent CuO_2 planes rotate in opposite directions [16, 19, 20]. Such continuous spin rotation can be induced by B because the spins gradually rotate toward a configuration perpendicular to the B at high fields. Therefore, the hard and easy spin axes are tuned by the field. A intriguing feature is the evolution of MR diagram with temperature. With decreasing temperature, the fourfold oscillations in type-I collinear phase are replaced by a twofold sine wave like feature at T_{N2} ; consequently a new fourfold oscillations show up in type-II phase, and are replaced by another twofold symmetric wave like feature across T_{N3} ; with further cooling fourfold symmetric oscillations develop. The MR diagram is fairly symmetric in type-I and type-III collinear phase, while is asymmetric in type-II phase. The MR diagram rotates by 45° in the type-II collinear phase relative to the type-I collinear phase, and the MR diagram with twofold symmetry at T_{N2} and T_{N3} rotates by 90° each other. Surprisingly, this is quite consistent with the spin reorientation from Phase I to II (all the Cu spins rotated by 90° about the c-axis). However, it is strange that the MR diagram does not change from type-II to type-III collinear structure. So the maximum MR is observed with B along Cu-O-Cu direction in type-II and type-III collinear structure, while with B along Cu-Cu direction in type-I phase. This is consistent with the magnetic hysteresis in MR observed

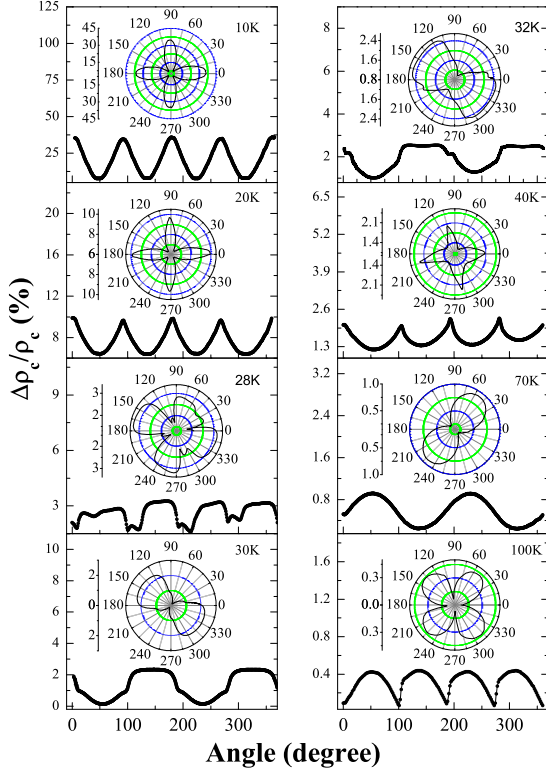


FIG. 4: The out-of-plane MR as a function of angle for $Nd_{1.975}Ce_{0.025}CuO_4$ crystal at different temperatures and polar plot of the same data.

in Phase II and no magnetic hysteresis in MR observed in Phase III. These results should be closely related to the Nd and Cu ion interaction, so that the MR shows different behavior for the same Cu spin order in phase I and III. It suggests that the MR effects are quite sensitive to the differences in the different collinear spin structures.

It should be pointed out that no MR anomaly and no hysteresis behavior in MR and in $\rho_c(T)$ are observed when a c-axis aligned field is applied, consistent with no transformation from noncollinear to collinear spin structure for such field orientation[21]. Compared to the lightly doped Pr-Ce-Cu-O material, the MR behavior is similar to each other due to the transition from non-collinear to collinear spin structure. However, the unique feature is the thermal hysteresis in $\rho_c(T)$ and the magnetic hysteresis in MR. The thermal hysteresis at spin reorientation transition cannot be observed at zero field $\rho_c(T)$, suggesting that the field has effect on the interplanar interactions since the spin reorientation transition arises from the competition of the various interplanar interactions. It is the effect of B on various interplanar interactions to lead to the FC MR behavior different from the ZFC MR case as shown in Fig.3. In addition, the evolution of the MR diagram with the temperature shown in Fig.4 is consistent with the spin structure transition at T_{N2} . The maximum MR appears with B along Cu-O-Cu direction in type-II collinear phase, while with B

along Cu-Cu direction in type-I collinear phase. It suggests that the hard and easy spin axes are different in type-I and -II collinear spin structures. Which could be the origin for the different MR behavior between FC and ZFC processes shown in Fig.3. The MR diagram does not change across T_{N3} , so that no magnetic hysteresis in MR is observed. But a remarkable thermal hysteresis is observed at T_{N3} . It suggests that the thermal hysteresis observed in Fig. 2 has a different origin from the magnetic hysteresis in MR shown in Fig.3.

In summary, the transport properties and the MR behavior are systematically studied in antiferromagnetic $Nd_{2-x}Ce_xCuO_4$. The transport properties is very sensitive to the subtle changes of the spin structure. We give a direct evidence for the itinerant electrons directly coupled to the localized spins. The thermal hysteresis in $\rho_c(T, B)$ and the magnetic hysteresis in MR are found. The hysteresis arises from the effect of the field on the interplanar interactions, such as: coupling between Cu and Nd ions, and the different hard and easy spin axes in the collinear spin structures.

Upon preparing this Letter, we became aware of a similar hysteresis observed in neutron scattering and MR experiments for $Nd_{2-x}Ce_xCuO_4$ [22].

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